



Comparative geochemistry of four ferromanganese crusts from the Pacific Ocean and significance for the use of Ni isotopes as paleoceanographic tracers

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Abstract

Ferromanganese (Fe-Mn) crusts are potential archive of the Ni isotope composition of seawater through time. In this study we aim at (1) understanding Ni isotope fractionation mechanisms and metal enrichment processes in Fe-Mn deposits, (2) addressing global vs. local control of Ni isotope composition of these deposits. Two Fe-Mn crusts from the North Pacific Ocean (Apuupuu Seamount, Hawaii) and two Fe-Mn crusts from the South Pacific Ocean (near Rurutu Island, Austral archipelago of French Polynesia) were characterized for their elemental geochemistry and Ni isotope composition. Geochemical analyses were performed at millimeter intervals in order to provide time-resolved record of Ni isotopes. Chronology and growth rates were determined using cosmogenic ¹⁰Be isotope abundances. The results show that, despite different growth rates, textures and geochemical patterns, Fe-Mn crusts from both North and South Pacific Oceans have fairly homogenous Ni isotope compositions over the last ~17 Ma, yielding average $\delta^{60/58}\text{Ni}$ values of $1.79 \pm 0.21\%$ (2sd, $n = 31$) and $1.73 \pm 0.21\%$ (2sd, $n = 21$) respectively. In one crust sample, however, layers directly in contact with the altered substrate show anomalously light $\delta^{60/58}\text{Ni}$ values down to $0.25 \pm 0.05\%$ (2se) together with rejuvenated ¹⁰Be/⁹Be ratios correlating with elevated Ni/Mn ratios. Such patterns are best explained by protracted fluid–rock interactions leading to alteration of Mn-phases after crust formation. Isotopically light Ni would be the result of Ni isotope fractionation during adsorption rather than the contribution of external Ni sources (e.g. hydrothermal sources) having light Ni isotope compositions. The combination of our results with previously published data on Fe-Mn crusts indicates that the average Ni isotope composition in deep waters has not changed through the Cenozoic (~70 Ma). We propose that Ni isotope variations in Fe-Mn crusts may not only record variations of Ni sources to the oceans, but also post-depositional processes depending on the growth history and geological settings of Fe-Mn crusts.

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1. INTRODUCTION

Fe-Mn crusts are seafloor metalliferous deposits forming through slow accumulation of seawater-derived Fe- and Mn-oxyhydroxide colloids on hard substrates such as volcanic seamounts that are kept sediment-free for millions of years (Craig et al., 1982; Halbach et al., 1983; Hein et al., 1988, 1992; Koschinsky and Halbach, 1995). Fe-Mn crusts are particularly abundant in the Northwest Pacific, e.g., the Pacific Prime Zone (Hein et al., 2013) and in the South Central Pacific (e.g., French Polynesia; Kosakevitch, 1987; Pichocki and Hoffert, 1987; Lesuave et al., 1989; Kosakevitch and Disnar, 1997) in connection with the formation and aging of Jurassic to Cretaceous volcanic seamounts (Halbach et al., 1983, 1984; Halbach and Puteanus, 1984; Aplin and Cronan, 1985; De Carlo et al., 1987; Hein et al., 1988, 1992). A seawater origin for trace metals incorporated into Fe-Mn crusts has been well documented using isotopic and trace element geochemical signatures including rare earth elements (Bonatti and Joensuu, 1966; Bonatti et al., 1972; De Carlo et al., 1987; Nicholson et al., 1997; Hein et al., 2012; Bau et al., 2014). The importance of oceanographic parameters such as the depth of the oxygen-minimum-zone (OMZ), the calcite-compensation-depth (CCD) and phosphatization events that occurred during the Cenozoic have been well-recognized to influence Fe-Mn crusts geochemical composition (Hein et al., 1993). Hydrothermal input of metals have been also suggested in some cases (van de Flierdt et al., 2004; Chu et al., 2006; Horner et al., 2015), which is in line with the recognition that hydrothermal venting may impact regionally the inventory of metals, in particular Fe, in the deep ocean (Toner et al., 2009; Tagliabue et al., 2010; Conway and John, 2014; Fitzsimmons et al., 2014). In rare cases, cosmic spherules were also reported in South Pacific crusts (Kosakevitch, 1987; Halbach et al., 1989; Lesuave et al., 1989; Kosakevitch and Disnar, 1997) but it remains unclear whether such extra-terrestrial input could affect bulk Fe-Mn crusts composition.

Radiogenic isotope geochemistry (Pb, Sr, Nd, Os, Hf) in Fe-Mn crusts has been extensively investigated in order to resolve fundamental paleoceanographic processes such as the evolution of oceanic circulation resulting from opening or closure of oceanic passages (e.g. closure of the Panama gateway, opening of the Drake passage), modifications in continental erosion fluxes after major climatic changes (von Blanckenburg et al., 1996b; Abouchami et al., 1997; Ling et al., 1997, 2005; O’Nions et al., 1998; Reynolds et al., 1999; Frank, 2002; Frank et al., 1999, 2002; van de Flierdt et al., 2003; Peate et al., 2009; Chen et al., 2013), and the impact of hydrothermal inputs (van de Flierdt et al., 2004). It should be noted that most of these studies were carried out on samples from the North Central Pacific Ocean. However other oceanic regions, including the South Pacific, have also important Fe-Mn crusts deposits (Hein et al., 2013) and they are poorly documented.

Stable isotope composition of metals (e.g., Fe, Zn, Ni, Cd, Cu, Tl, Mo) in Fe-Mn crusts and nodules, provide interesting proxies to unravel changes in the marine sources and biogeochemical cycles of metals through time (Zhu

et al., 2000; Barling et al., 2001; Rehkämper et al., 2002, 2004; Siebert et al., 2003; Levasseur et al., 2004; Anbar and Rouxel, 2007; Schmitt et al., 2009; Horner et al., 2010, 2015; Nielsen et al., 2011). However, the application of Ni stable isotopes as paleoceanographic proxies is not straightforward since at least three processes may affect the Ni isotope records preserved in Fe-Mn crusts: (1) changes in the relative fluxes of Ni sources and sinks in the ocean, (2) internal biogeochemical cycling of Ni and water mass mixing, and (3) fractionation processes during Ni incorporation at the Fe-Mn crust surface (Wasylenki et al., 2008, 2011, 2014a,b; Gueguen, 2013; Nielsen et al., 2013; Bryan et al., 2015) and Ni distribution among Mn-phases (Koschinsky and Halbach, 1995; Koschinsky and Hein, 2003).

The isotope composition of Ni in marine systems has recently attracted significant attention (Cameron and Vance, 2014) owing to its nutrient-type behavior (Sclater et al., 1976; Cameron and Vance, 2014) and multiple sources in seawater (e.g., rivers, atmospheric depositions, hydrothermal vents; Gall et al., 2013; Cameron and Vance, 2014; Little et al., 2015). In a reconnaissance study, Gall et al. (2013) reported up to 1.6‰ variations in $\delta^{60/58}\text{Ni}$ ($\delta^{60/58}\text{Ni} = ({}^{60}\text{Ni}/{}^{58}\text{Ni}_{\text{sample}}/{}^{60}\text{Ni}/{}^{58}\text{Ni}_{\text{NIST986}} - 1) \times 1000$; see Section 2.3) along the growth layers of Fe-Mn crusts from various ocean basins. The apparent absence of relationship between Ni isotope variability and sample locations and depths implies that differences in water masses, surface water productivity and bottom water oxygenation have no influence on Ni isotope signatures preserved in Fe-Mn crusts. The Ni isotope composition of surface scrapings of Fe-Mn crusts also shows significant variations in $\delta^{60/58}\text{Ni}$ values (from 0.9‰ to 2.5‰) with values being both heavier and lighter than the modern seawater value determined at $1.44 \pm 0.15\%$ (Cameron and Vance, 2014). While it has been argued that heavier Ni isotopes values may be due to continental input and lighter Ni isotope values reflecting the extent of hydrothermal input in deep waters (Gall et al., 2013), it remains unclear which processes are controlling Ni isotope record in Fe-Mn crusts. Presumably, the importance of local or post-depositional effects associated with Ni adsorption and precipitation in Fe-Mn crusts should also be considered as a potential cause of isotope fractionation as suggested by experimental results of Ni sorption on Fe-oxyhydroxides (Sorensen et al., 2011; Gueguen, 2013; Wasylenki et al., 2015) and Mn-oxyhydroxides (Gueguen, 2013; Wasylenki et al., 2014a) indicating enrichment of the mineral phase in light Ni isotopes, keeping in mind that Ni is essentially hosted in the Mn-phase.

Here, we report Ni isotope composition of two pairs of Fe-Mn crusts collected on two volcanic seamounts from the Northern Pacific Ocean (Apuupuu Seamount, Hawaii) and the Southern Pacific Ocean (near Rurutu Island, Austral archipelago of French Polynesia). This approach allows (a) a direct comparison of Ni isotope record in Fe-Mn crusts from the same seamount in order to address local effects, and (b) a comparison of geochemical composition of crusts between North and South Pacific in order to address the effect of more global geochemical processes.

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